Problems in Evaluating Radiation Dose via Terrestrial and Aquatic Pathways

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This review is concerned with exposure risk and the environmental pathways models used for predictive assessment of radiation dose. Exposure factors, the adequacy of available data, and the model subcomponents are critically reviewed from the standpoint of absolute error propagation. Although the models are inherently capable of better absolute accuracy, a calculated dose is usually overestimated by from two to six orders of magnitude, in practice. The principal reason for so large an error lies in using "generic" concentration ratios in situations where site specific data are needed.

Major opinion of the model makers suggests a number midway between these extremes, with only a small likelihood of ever underestimating the radiation dose. Detailed evaluations are made of source considerations influencing dose (i.e., physical and chemical status of released material); dispersal mechanisms (atmospheric, hydrologic and biotic vector transport); mobilization and uptake mechanisms (i.e., chemical and other factors affecting the biological availability of radioelements); and critical pathways. Examples are shown of confounding in food-chain pathways, due to uncritical application of concentration ratios. Current thoughts of replacing the critical pathways approach to calculating dose with comprehensive model calculations are also shown to be ill-advised, given present limitations in the comprehensive data base. The pathways models may also require improved parametrization, as they are not at present structured adequately to lend themselves to validation. The extremely wide errors associated with predicting exposure stand in striking contrast to the error range associated with the extrapolation of animal effects data to the human being.

Introduction

This review is concerned with radiation dose assessment and the accuracy with which it determines the calculated risks of incurring health effects from exposure to radiation. Radiation dose estimation, as used for assessment purposes, depends critically on how exposure factors are determined. We are not here concerned with estimating human health effects by extrapolation of animal data to humans. Such an extrapolation has been treated elsewhere (1).

Several prevalent misunderstandings about the significance of radiation dose estimates, and about

more general applications of this approach, have suggested a need for comprehensive review of dose estimate methods. Those responsible for performing dose assessments have usually been well aware of constraints limiting their accuracy (2). However, many other persons concerned with dose data, in the biomedical, engineering and legislative communities, have believed that the mathematical models used to generate the dose estimates are capable of a degree of accuracy that is now impossible to achieve in practice. Rather few people have noted that the model developers, themselves, have believed the absolute errors in the use of their models led to doses overestimated by anywhere from two to six orders of magnitude (2, 3). As contrasted with uncertainties from two-fold to ten-fold in the prediction of human

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health risks from laboratory data (1), it is therefore highly desirable to refocus attention on the models used to estimate dose and the factors used to integrate these estimates into projections of human health effects.

When calculated annual radiation doses were often only a small fraction of then allowable limits, inaccuracy of dose estimates although large was not so important a consideration. Subsequent developments have changed this situation drastically; i.e., issuance of FRC guidelines in 1960, and particularly the gradual development of as-low-as reasonably-achievable (ALARA) philosophy. At the same time environmental concentrations resulting from human endeavors declined for the most part to below natural background levels. With the application of ALARA, a convergence of radiation standards and environmental concentrations has occurred. The response to this has been to attempt to improve accuracy in calculating dose with increased model sophistication; i.e., inclusion of more exposure pathways of increased complexity. Yet, the need is to improve the quality, specificity, and realism of both model parameters and data. A more judicious selection of existing data, some additional research, and a reconsideration of present model structure are, all, involved.

We will examine here some key considerations toward improving the quantitative usefulness of exposure models. Both model structure and specific problems in the data base will be evaluated.

Characteristics of Exposure Models

Several mathematical models, and the computer codes that implement them, have been described in detail elsewhere (4-7). Many of their elements are provisional, but the models provide the only practicable way of accounting for an extremely large number of variables (see Fig. 1). Such models have utility where the substance under study (1) is comparatively stable, (2) is noxious at low levels, (3) disseminates through a multiplicity of environmental pathways and (4) requires comparatively long periods for tissue accumulation and induction of effects. The most serious constraints in using the models are the needed development of a sometimes impracticable data base required for implementation, and the comparative lack of data specific to real locations.

When measurable concentrations of radionuclides in air, water and foods existed near large AEC installations, it was a simple matter to calculate radiation dose by combining the measured concen-

trations with living and dietary habits. The calculated annual doses were generally a small fraction of the then existing limits, with only a few exceptions. In addition, radioactive fallout from nuclear weapons testing provided measurable concentrations of several specific radionuclides in the environment, especially ^{89,90}Sr, ^{131}I and ^{134,137}Cs. These two sources of environmental radioactivity provided some of the first (and in certain instances the only) field data on the behavior or radionuclides in the environment. They also, quite naturally, gave rise to empirical equations designed to predict the concentration of selected radionuclides in human diet (8, 9).

To consider a cattle grazing ecosystem, for example, the relevant compartments would include air, water, soil, primary producers (plants), primary consumers (grazers), secondary consumers (predators) and decomposers (bacteria, fungi), any of which can be further divided, e.g., grasses, forbs, shrubs and trees for the primary producers. For food chain evaluation, the requisite level of detail will depend on who is eating and what is being eaten specifically (10).

Additional model subcomponents are needed to deal with variabilities in dietary consumption patterns (critical populations), living habits (workplace, home, outdoors), and the specific physiology of given radioelements (assimilation, retention, critical organs). The necessary computer subroutines are well established in current codes for computing radiological dose (7, 11, 12). They are under more or less continuous revision. An early ICRP publication (13), its numerous subsequent publications, and the deliberations of model builders should be consulted for current details (3). The present discussion will be limited to environmental pathway subcomponents, now in need of better focused data.

Depending on regional land use, cultural or other considerations, the subcomponent models may have to be examined for food chains different from the major agricultural chains on which the food preference models are usually built. Recreational hunting of deer or subsistence fishing can be of regional political and social significance, for example, but in general, data are sparse for non-agricultural food chains. These chains will usually require site-specific data acquisition.

The modeling problem is one of determining the distribution of pollutant concentrations in a compartment. Functional connections between compartments are thus represented by steady state interchanges. Typical linkage processes include adsorption, absorption, inhalation, ingestion, excretion, decomposition, and dissolution. Few of
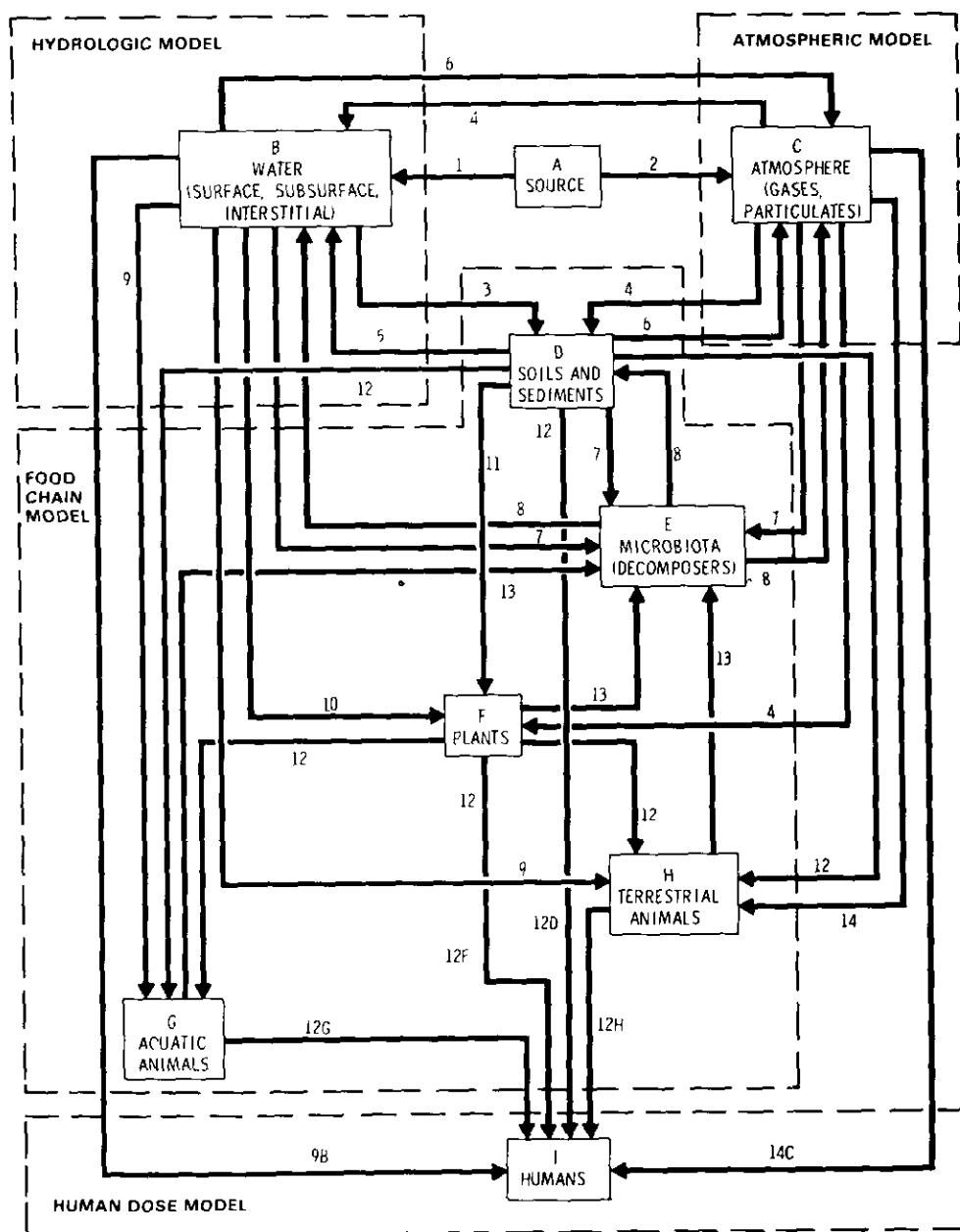
FIGURE 1. Environmental pathways model for dose assessments showing major routes affecting man: (1) aquatic discharge; (2) atmospheric discharges; (3) irrigation, water/sediment interphase exchanges; (4) surface deposition; (5) leaching, erosion, mineralization, sediment/water interphase exchanges; (6) resuspension/evaporation; (7) microbial incorporation; (8) microbial releases, decomposition; (9) skin absorption; (10) irrigation plant surface exchanges; (11) root uptake; (12) ingestion; (13) mortality; (14) inhalation. The representation above delineates relationship between 4 major submodels, on which dose assessment depends. The detailed computer codes are too complicated to permit diagrammatic representation and no one submodel is adequate by itself to determine critical pathways (see text).

these processes follow the reaction kinetics, in fact, assumed by the model builders. It is clear that seasonal production (vegetation) and reproductive cycles affecting population size (fish, animals and birds) lead to modeling difficulties because of decidedly non-steady state conditions (14). Our current inability to appropriately describe these time dependent processes sometimes leads

to excessive variability in projecting dose estimates, and it certainly prevents application of the present modeling approach to predicting ecosystems responses. Several attempts at rigorous compartment modeling have been only partially successful (14-20). More work is needed also on the parameterization of transport rates.

Source Considerations Influencing Dose

Particle size, physical state, molecular form, the presence of codisposed organic complexes, and release rates are basic factors controlling biological availability of a radioelement to different biotic receptors. Since data of these kinds are costly to obtain, process engineers seldom provide them. If they are to be provided, some selectivity is required of the biologist. The biologist should prioritize the needed information, based upon an understanding of the metabolic potential of released materials.

Among the factors indicated above, the biological importance of aerosol particle size is often overlooked. In terrestrial ecosystems, both particle form and size determine subsequent soil/root behavior and interaction on the plant leaf. This is true whether the particle originates in the upper atmosphere or from wind-resuspended soils. Suitable particle size data are rarely available to interpret observed plant uptake of radioelements. As a general rule, the smaller the size of particle deposited on a plant, the greater its likelihood of biological interaction, regardless of its chemical solubility. A leaf can sorb and translocate components of initially anhydrous compounds and compounds of extremely limited solubility (21). Particles of 1 μm or less in size are unlikely to be washed off the leaf (22), a point not generally appreciated. Adherence is probably related to epidermal structures, the surface area/volume ratio, and charge densities on the surface macromolecules of the leaves. Some particulates in air, like sodium-plutonium oxide and certain refractories, are unusual in disintegrating on contact with water to form exceedingly small particles (23). It is not known how readily assimilable these smaller particles may be, although in animal body fluids, translocation is enhanced 45-fold (24). There are very few nuclear industrial data on size of the particulates released and no data on how particulates are transformed in the atmosphere. The EPA requires measurement of the number of suspended particles, but it does not currently require the determination of the particle size distribution (25).

In aquatic ecosystems, particle size governs absorption and feeding behavior. Generally, particles less than 100 μm size carry a significant fraction of the radioactivity. Below this size, they are suspendable and ingested by filter feeders, and subsequent salinity increases in estuarine situations can promote movement of a radioelement off the particle to the water from which it may be more readily absorbed. These processes are well recognized (26), but the kind of particle chemical and physical data necessary to predict or interpret organism uptake are exceedingly rare (27).

A similar problem exists regarding the molecular forms of released radioactive contaminants. One of the more complete documents giving information on source terms is the LMFBR Final Environmental Statement (28). For the nuclear fuel cycle, excluding uranium mining, nineteen radioelements were identified as potentially of concern because of either high toxicity in biological tissues or high comparative release rates. There was no information on molecular form available in this report. While biologists generally assume that the environmental behavior of radioactive elements will be representative of the simple chemical forms (oxide or nitrate), this can be misleading. Environmental behavior of certain long-lived gaseous radioelements, like iodine, is often predicated on elemental chemistry, although methyl iodide and other alkyl iodides up to a C_8 chain length have been identified as principal forms that may escape from certain process vessels during nuclear fuel reprocessing (29). Certain elements, when bound to organic ligands produced by metabolism, are more biologically available than they are in inorganic forms (30). This is particularly the case for plutonium (31). Codisposal is a related concern. Radioelements complexed inorganically or forming chelates may be more or less biologically available, depending on the metabolic characteristics and specific binding kinetics of the complex molecule. Significant amounts of complexing agents are used in the nuclear fuel reprocessing industry (Table 1), as they are in agriculture. In the nuclear industry, the extent of release to the environment is not always clear (32, 33).

The environmental and metabolic behavior of technetium, a potentially large contributor to radiation dose, has been widely investigated in recent years. In 1959 the ICRP (13) defined the metabolic behavior of Tc and indicated that it was taken up less readily than iodine. Nevertheless, nuclear engineers considered it to be an iodine-like element. The use of the pertechnetate form of Tc in thyroid function (uptake) tests as a substitute for radioiodine began a few years ago. Since then, the

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Table 1. Chemical consumption in a nuclear fuel reprocessing plant.[a]

Chemical	Consumption, lb/yr
Phosphoric acid	3,000,000
HEDTA	2,000,000
EDTA	1,500,000
Hydroxyacetic acid	550,000
Citric acid	300,000
Sodium glutamate	170,000
Tributyl phosphate	129,000
Oxalic acid	35,000
Di(2-ethylhexyl)phosphoric acid	35,000
Phosphotungstic acid	22,000
Polyacrylamide	6,300
Tartaric acid	6,000
Dibutyl phosphate	3,500

[a]Taken from ERDA (*32*).

possibility that technetium-99 exists in the environment in the pertechnetate form has stimulated several studies of its form and uptake by plant roots (*34*). Several researchers also reported surprisingly high Tc uptake by plants from soil based on laboratory experiments (*35, 36*). All of this led to heightened concern that the potential doses from Tc-99 in the environment were being grossly underestimated. However, more recent research has indicated that the appropriate concentration ratio (CR) for use in the food-chain models are lower than first implied by the laboratory studies and that Tc ingested with food does not behave like iodine in the body (*37*). This situation illustrates that additional research may need to be done on several other elements where chemical similarity to another element was assumed for modeling purposes.

Dispersal Mechanisms

In the terrestrial environment, atmospheric dispersal/deposition, direct discharge to waterways, and leachates from soil-stored wastes are governing pathways for soil and water concentrations. The first principles of these processes are fairly well understood, and it is mainly the specific interactions of particular elements that present problems. Airborne materials that deposit directly onto surface waters usually make a small contribution to water concentrations compared to material deposited onto land in the watershed and later washed into the river or lake (*38*).

Atmospheric Dispersal

Because people and green plants can act as integrators over time for the low level, long-period

release of radioelements, those concerned with foodchain assessment are usually interested in annual mean deposition rates about the point of release. Several atmospheric models have been constructed for the prediction of annual deposition of trace metals from combustion facilities and for radioelements applicable to nuclear fuel cycle facilities, principally for open terrain (*38, 39*). These submodels, in Figure 1, have not been validated for deposition in forest or field canopies, and deposition is probably underestimated by their use. Recycling of radioelements from soil deposits to the green leaf (via wind resuspension, or gaseous diffusion from soil) also has not been measured. However, resuspended soil would appear to provide a 5- to 10-times greater source to the plant than the root/soil interface (*40*), and these data should be established.

Other unknown factors affecting the atmospheric submodel include: localized variations in deposition due to differences in type and intensity of precipitation, airborne particle properties and concentration, changes in deposition caused by air currents and eddies near obstructions, variable deposition induced by wind-shifts, deposition of large particles that cannot remain airborne, runoff and puddling of rainwater, and increased accumulation on polar surfaces. Present knowledge of the detailed deposition mechanisms for a specific terrain is neither adequate to predict local deposition, nor to know whether a geometric factor of 3 or some larger value brackets the range of accuracy for a calculated deposition. Need exists to integrate for validation purposes biological sampling with atmospheric dispersal tests, in order to circumvent the difficulties posed by these numerous processes.

Hydrologic Dispersal

Hydrologic dispersal involves several processes, the relative importance of which will depend on site-specifics. The processes include hydrodynamic transport, sediment sorption/desorption, sediment transport, and biotic transport. Of these, only hydrodynamic transport can be modeled with acceptable accuracy (geometric factor of 2 or smaller), and the models have been reasonably validated for lakes, rivers and impoundments (*3*). Different models are required for free-flowing water (river or stream) or standing water (lake or pool). Since most radioactivity of terrestrial origin is carried on translocated soil particles of small size, stream concentrations are controlled principally by particle size, which affects residence time (measured in months and years), and sedimentation rate. Only limited data are available (*27, 38,*

41). Many contaminants, found to be more concentrated in the sediments than in the water, may remain available to biota over long periods of time. Estimates of their biological availability from sediments can be made, in appropriately designed experiments (*42*, *43*), but this has been seldom done for radioelements.

Biotic Vector Transport

This process refers to the physical transport of radioactive material by animals or sometimes by wind distribution of plant materials (*44*). Food chain transfer is considered separately below. Biotic vector transport processes are believed to move trivial amounts of contaminants through the biosphere compared to atmospheric and hydrologic transport processes. However, this statement can be shown to be invalid for specific special situations. Also, the unexpected appearance of radiocontaminants in biota outside of the site boundary of a facility has led to significant, adverse public reaction. Typically these are situations where animals act as vectors for the dispersion of buried or stored radioactive material beyond its control point. Biotic transport processes have not generally been considered systematically. They are invariably site- and situation-specific processes and they include such diverse efforts as estimating off-site transport of surface buried low-level waste by deer, or estimating the amounts of low-level radioactive material exported from pond sediments and water by various waterfowl using a pond (*44*, *45*). Figure 2 and Table 2 show an instructive example where biomass and its seasonal variation were taken into account to establish upper and lower limits for the export routes.

At high exposure levels, biotic transport can represent a significant link in an environmental transfer pathway involving organism-to-organism food chains. In cases like this it becomes necessary to evaluate that entire pathway relative to other pathways (see below). When biotic transport is the governing mechanism, measuring its contribution to dose will require specific data on feeding habits of the organism, population size, migratory range, and relation to human habits (*44*). The first three kinds of data are primary variables affecting dose. They are measurable and can be determined with statistical reliability using specialized approaches. Metabolism of radioactive compounds transported by biotic vectors is sometimes important and may need to be considered.

Mobilization and Uptake Mechanisms

Biological Availability: Chemical Considerations

Only a very small fraction of radioelements bound to soils or aquatic sediments are available to biota, but this fraction is variable over a wide range. The concept of biological availability rests on several lines of evidence, but it has been systematized only in the soil/plant system: i.e., (1) ligand formation enhances plutonium uptake and translocation in plant and animal systems (*31*); (2)
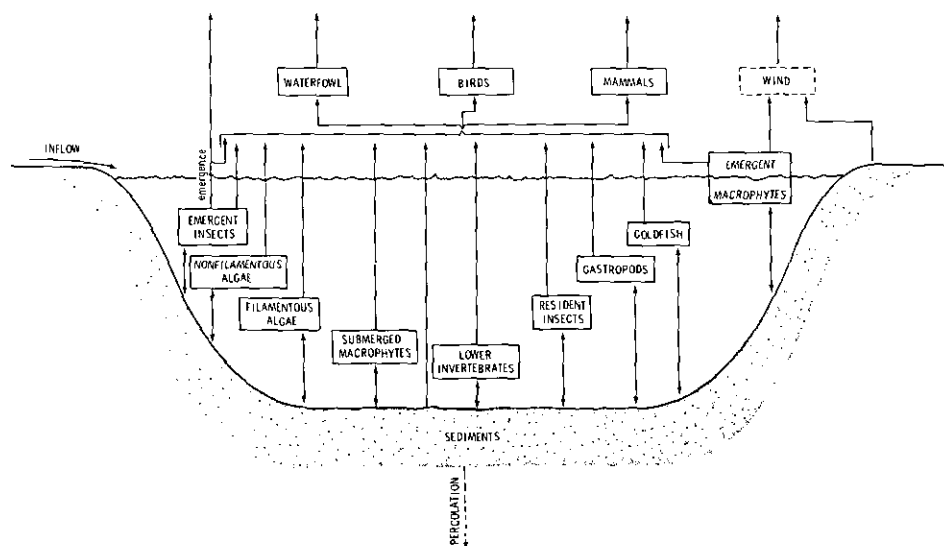

FIGURE 2. Export routes of radioactive materials from contaminated pond sediments.

	Pu, nCi
Inventory of Pu in	
Sediments	10^9
Diatoms and pondweeds	10^7
Emergent insects	10^4
Other fauna	10^5
Export of Pu by	
Emergent insects	10^4/yr
Resident waterfowl	10^1–10^3/yr
Other birds	<10^1–10^3/yr
Mammals (including deer)	10^1/yr
Wind	negligible
Percolation	negligible

the small fraction of heavy metals in soils that is actually exchangeable closely reflects soil amendment fractions (46); and (3) the factors controlling availability are both exogenous and endogenous. Exogenous factors may involve such source term variables as valence state of the radioelement, its molecular form, concurrent presence with chelates or other exogenous complexation agents, and particle size, as discussed earlier. Endogenous factors may involve ligand metabolites of soil microflora and plant roots, and geochemical factors determined by soil characteristics. Similar processes undoubtedly operate in sediment/microflora/detritivore systems of lakes and streams. Related processes may operate also in organism-to-organism transfer of plutonium through sequential food chains; e.g., gastrointestinal absorption of ligand-bound plutonium is significantly enhanced over the inorganic forms of plutonium subject to hydrolysis (30).

At present, the chemical basis for biological availability deals mainly with plutonium (31) and some heavy metals. One may expect that similar processes will be common to any radioelement whose chemistry is primarily controlled by hydrolysis or whose electron configuration favors complex ion formation. Among the long-term or large radiological contributors to dose this includes isotopes of uranium, plutonium and americium in the actinide series; strontium, cesium and cadmium; and nickel, iron and cobalt in the transition series. Complexation with soluble ligands may maintain the solubility of several of these elements, and thus compete with their sorption to soil and sedimentary particles. Such reactions do not necessarily increase their biological availability from the water column; i.e., larger complex ions may not be biologically absorbable whereas the small ones may be. Clearly, a great deal depends on the

type of polymer or complex ion formed, which in turn is a strong function of chemical and biological properties of particular bodies of water. These properties have been characterized in only a rudimentary way. For example, plutonium concentration in the water column of Canadian and Great Lakes was observed to vary 100-fold among lakes (47). Complex ion formation associated with high carbonate content or organic ligands and valence state associated with acidic waters evidently accounted for the differences (31).

In marine ecosystems, solution chemistry is fairly well-explained for plutonium (31). Little comparable information is available on other radioelements that might be significant dose contributors. Nevertheless some insight can be gained from nonradiological studies currently in progress; e.g., copper and cadmium form complexes with organic and inorganic ligands when added to seawater, and these molecular forms may significantly reduce their biological availability, as long as the ligand capacity of the water is not exceeded (48-50). Other evidence suggests that microfloral activity in the sediments is responsible for mobilizing copper (41-43, 51).

Biological Availability: Related Factors

The concept of biological availability has not always been restricted to purely chemical or microbial processes. Other factors that affect mobilization of a radioelement from a soil or sediment particle include uptake route, metabolism, and resuspension.

In aquatic ecosystems, radiocontaminants may enter by several routes and move with either the water, sediment, or biota. Numerous processes, not all of which have been delineated, affect each medium; e.g., detritivore and microfloral metabolism, pH and saline chemical changes, settling/sedimentation rates (52, 53), and sediment transport. It is probably impracticable to assess organism uptake as a function of all of the variables mentioned, and for this reason, recourse is had to concentration ratios, as actually observed at contaminated locations. However, for predictive purposes, this may not always be satisfactory because concentration ratios are often confounded as to routes of uptake (see following discussion). Of the various processes operating in the aquatic environment, perhaps most important are the development of research approaches to measure radiocontaminant movement from benthic sediments to their associated biota as discussed above.

In terrestrial environments, the situation is somewhat better delineated. For example, air-

borne radioactivity can enter plants via direct deposition onto the leaves, via root uptake or via deposition of resuspended material from soil to plant surfaces. In practice, each of these processes is often confounded, in the concentration ratios selected for the model. However, the relative importance of each mechanism can be estimated since measured values have been obtained for each. Relative importance will depend on the specific circumstances of the radioelement release.

Concentration Ratios

The concentration ratios widely used in dose-assessment models are significantly influenced by biological availability processes and uptake routes. The ratios themselves represent operational definitions that, e.g., in the case of plant/soil ratios, lump together soil desorption, root uptake, foliar uptake, microbial and soil solution chemical equilibria. Where the published concentration ratios vary over four or five orders of magnitude (3, 46, 54), each of these factors may need to be evaluated. This is equally true in aquatic ecosystems, where for example water to fish concentration ratios for radioactive cesium and strontium range over three or four orders of magnitude (3, 55, 56). Here, again, various processes are lumped together, water, sediment-water interaction, changes with chemical form, nutrient interaction, temperature effects, partitioning effects (physical form), biotic (vector) transport in detritivores, and metabolism. Of these factors water chemistry and sediment-water interactions appear to be most important (see preceding section). In the case of radiostrontium, the concentration ratio is inversely correlated with the calcium content of the water; for radiocesium it is inversely correlated with the potassium content of the water (57, 58).

In marine environments, the relative constancy in ocean water composition has led to the belief that water to organism concentration ratios are well established. However, ocean discharges are made at shoreline into waters subject to fresh water runoff and the influence of man's activities. Recent research suggests that concentration of trace elements in coastal water is highly variable, particularly for non-conservative elements. The kind of tabulations in which concentration ratios are averaged for laboratory and field data therefore would probably underestimate radioelement uptake in estuarine and some coastal waters (3, 59). This may not be true where directly observed field measurements are used, as for the Cap de la Hague and Windscale studies (60).

Second, the values for many radioelements have not been measured but have been inferred by chemical or biological analogy to other elements. Third, for some radioelements only a very limited number of measurements exist and these values are seldom accompanied by the necessary description of the experimental conditions which possibly affected the results. In some instances the experiments were conducted under inappropriate conditions; e.g., potted plants were grown in a small enclosure rather than under open field conditions; the plants were not taken to maturity, the specific plant grown or the parts of the plants analyzed were not those usually consumed; and, the chemical forms of the radionuclides used may not have existed in the terrestrial environment.

Over the past five years, notable progress has been made in delineating the factors accounting for such variations as those indicated above. However, radiation exposures are calculated every day, using standardized tables of data (4, 59, 61) in which the propagation of errors at each link in a complex chain might lead to a large cumulative error in the end (54). As the ratios in use are without intrinsic physiological significance, there can be no one "correct" value for all circumstances. Currently used ratios only provide a basis for generic pathway calculation, with its attendant large error. Data developed and selected for similar sites would definitely provide more accurate calculations.

Critical Pathway Considerations (Food Chains)

Referring again to Figure 1, the concept of critical pathways has been used to permit needed simplification of an already too complicated environmental model (62, 63). If calculation using generic concentration ratios leads to minor dose contributions from a radioelement being transported in certain pathways, these pathways might be feasibly ignored. Attention then might be focused on getting specific data to determine transport in the dominant pathways. Today, there is discussion of abandoning this approach for regulatory purposes, instead attempting a total mathematical model computation. Considering the many sources of error outlined in the foregoing sections, and the provisional nature of much of the present radiological data base, this choice is ill-advised.

The importance of critical pathway evaluation will be shown. For example, one can demonstrate that the foliar uptake of airborne plutonium is a much more important pathway than root uptake

from human dose assessment. Using the computer code, FOOD (12), we computed the concentration of radionuclides in and on vegetation (and the subsequent radiation dose contributions following ingestion) via foliar deposition separately from root uptake. Inhalation doses were calculated using the DACRIN computer code (64). DACRIN is based on the ICRP Task Group Lung Model (65). Both inhalation and ingestion doses were calculated assuming the intake took place uniformly throughout one year. Sensitivity analysis performed with the food chain portion of the HERMES model (66, 67) showed the relative importance of the foliar pathway (up to 50 times the root uptake path) at least for releases to the atmosphere over a few years duration. Further calculations using food chain models (61) derived from HERMES yielded similar results after longer term (decades) accumulation in soil (68).

Table 3 shows the relative contribution to dose via each pathway for five cases incorporating several environmental and metabolic parameter values. In all five cases the same constant ambient air concentration was assumed to exist at the point of inhalation and at the place where the vegetation was grown. Results were expressed as percent of total dose in bone contributed by each of three pathways, i.e., inhalation, ingestion via the foliar deposition route and ingestion via the root uptake route. Similar values were obtained when total body was used as the organ of reference rather than bone. For this air release situation, the majority of the dose was of course from inhalation. In another release situation, we will discuss other routes that may be more important.

The relative contributions shown in Table 3 do not apply equally to all radionuclides. Use of codes structured to permit determination of dose contributions from individual nuclide or pathway (5, 11),

has indicated that radiation doses from ingestion of certain other radionuclides can be higher than those from inhalation given the same initial air concentration. The most obvious example is the radiation dose from radioiodine via milk consumption, with a somewhat smaller contribution via vegetables (68-70). Similar but less dramatic relationships exist for other fission products as pointed out by Garner (8). Even in these instances, however, the contamination reaching the food via foliar deposition outweighs that from root uptake even after many years of chronic deposition to soil (68).

The results just discussed were obtained using a fraction of 25% of deposited material initially on the plant (75% on the soil) and an exponential half-life for plant retention of 14 days (8, 71). This half-life and retention function have been experimentally determined for a few radionuclides, e.g., strontium and iodine (8, 9), but the retention function is undoubtedly more complex. Fractional interception by plants is a function of the density of vegetation cover, weather, and chemical and physical properties of the contaminant. Certain experiments have demonstrated that the 14-day half-life does not hold for several types of particles in the 1 μm range (22, 71). While directly applicable data are sparse, we believe that half-time on vegetation is a multiple exponential function with half-times for particulate contaminants that depend on particle size. Wind resuspension of contaminated soil also may lead to greater plant uptake through the leaf than through the root, but these observations have not been established on a quantitative basis. Recently, McPherson and Watson (72) estimated the added radiological impact of resuspension of plutonium deposited on soil during hypothetical accidents. Air concentrations and redeposition rates onto plant surfaces were calcu-

Table 3. Relative importance of the inhalation and ingestion routes in the calculated radiation dose to bone from plutonium for selected environmental and dietary parameter values.[a]

Case	CR value (dry plant/ dry soil)[b]	Diet, kg/yr	Fractional uptake (GI tract to blood)	Years of soil accumulation[c]	Proportion of total bone dose, %		
					Via inhalation	Via ingestion of vegetation contaminated by:	
						Foliar deposition	Root uptake
1	0.001	300	3×10^{-5}	1	99.	1.	2×10^{-7}
2	0.4	300	3×10^{-5}	1	99.	1.	8×10^{-5}
3	0.001	300	3×10^{-5}	51	99.	1.	1×10^{-5}
4	0.4	300	3×10^{-5}	51	99.	1.	4×10^{-3}
5	0.4	1000	3×10^{-4}	51	84.	16.	0.1

[a]All values have been rounded to no more than two significant figures.
[b]Data of Vaughan et al. (22).
[c]Deposition velocity set at 10^{-3} m/sec.

lated by numerically integrating the resuspension equation of Anspaugh (73) out to 50 years post-accident. The total resuspended Pu added about 80% to the initial ingestion dose resulting from direct deposition onto plants during the accident. They also calculated that about 80% of this total 50-year exposure to resuspended material occurred in the first year and 99% occurred in the first five years. Nearly all of the dose from ingestion of plant materials was accounted for by the first year's foliar deposition both direct and resuspended, and root uptake was negligible (less than 1%). Clearly, the quantitative implications of an aerial input to food chains may be different than that of a low-level burial input through soil. These differences need to be evaluated at specific sites.

Examples also can be found in freshwater ecosystems where high CR values for many radionuclides in aquatic foods make the ingestion of these items much more significant than the ingestion of drinking water.

Accuracy of Dose Estimates

The authors of the environmental support document to the Generic EIS for Commercial Waste Management (2) have given their thoughts on the subject:

"The consensus of those individuals contributing to dose assessments is that for any given dose estimate, the actual dose that would be received by the regional population in the reference environment would not be more than 10 times the stated value, nor would it be less than 1/100 of the stated value. Thus, the likelihood of actual values exceeding estimates is low, whereas the likelihood of values actually being substantially less than estimated is rather high."

"Because of additional uncertainties in modeling for a worldwide dose, the consensus is that doses received by the worldwide population would not be more than 100 times the estimate given, nor less than 1/1000 of the value given."

". . . doses presented in this report are best estimates of the doses; it would be improper to multiply all doses by 10 when it is just as likely that the true dose is 1/100 of the stated dose."

Some authorities have estimated that the absolute error band in calculated radiation exposure to a population exposed through aquatic food chains might be millionfold (3). Any of these statements have yet to be proven or disproven!

The solution was recognized in the RIME report where R. F.Foster state (26) "Perhaps the greatest uncertainty in predictive calculation is the

selection of an appropriate concentration factor . . ., [and] . . . wide variations that have been observed among different environments, and even among closely related species in the same environment emphasize the need for careful consideration of the specific characteristics of each site" (p. 247). Current practice is to use generic concentraleast, have been acceptable for routine determination of compliance (dose to maximum individual) when the computed doses represented a small fraction of the design limit. Health physicists are forced to make decisions on which parameter values are most applicable to their particular situation when sometimes there is only one value available. The value may have been determined under conditions different from those involved in the dose calculation. Such difficulties have led to the general practice of using parameter values which assure that the calculated radiation doses have not been underestimated. Maximum possible parameter values are seldom chosen for every step in the dose calculation, but whenever a parameter value is in doubt the tendency is to select a value that yields a conservative estimate (i.e., overestimates the dose). However, when the exposure standards are being lowered, more realistic data need to be obtained and used. The potentially large error in calculated radiation exposure represents a powerful incentive to obtain information and data applicable to a specific region or site.

Over the past decade, "generic models" have been used increasingly for evaluation of alternative sites, alternative nuclear fuel cycles, or alternative radioactive waste processing methodology. Except in the latter instance, calculated doses have seldom been the determining factor in the final choice. Economics, availability, and status of technology are often the prime selection criteria. This lack of impact on the final decision stems from the fact that comparisons are performed before detailed site-specific data are available or even sought. The belief exists, among those performing generic comparisons, that the effects of unknown or poorly known factors are often similar in each case so that the lack of knowledge does not seriously alter the final choice. One should note, however, that generic data when used for the model parameters reduce the chance that differences between alternatives will be evident. By contrast in site-specific evaluations, the regulators and the public potentially affected have come to insist that the dose estimates include all potential pathways of exposure and all necessary parameters, as realistically as possible.

Pathway analysis and radiological assessment must be continuing reiterative processes until

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there is sufficient confidence in both the mathematical models and the parameter values for unequivocal decision making. In fact, this second need has been recently recognized by Committee 64 of the NCRP, which is considering the problem of site-specific data.

Considering the above problems, particular caution also needs to be exercised in applying a set of concentration ratios derived from one set of exposure circumstances to a new set of circumstances. When comparing different situations, such as surface stored sources, fugitive emissions from industrial facilities, and transportation accidents, one finds that the relative importance of the several pathways shown in Figure 1 is different for each situation.

Goals and Future Direction

Current research on environmental pathways should be aimed at two goals, not readily achievable at the present time: (1) improving the absolute accuracy of dose estimation, and (2) validating the predictive capability of dose estimation models, by field measurements under controlled conditions. To accomplish these goals, particular attention needs to be paid to the following considerations.

● Accelerating the collection of site-specific data bases. This must be done for specific configurations of release, for specific molecular forms of the released material, and for specific physical and ecological features of the site.

● Developing generic quantitative approaches for those descriptive processes that have not yet been quantitated. This includes a determination of the range over which such variables may change.

● Improving the structure and parameterization, particularly, in the present dose assessment models. The models as shown in Figure 1 are not adequately structured to lend themselves readily to validation. More rigorous evaluations are needed to investigate model sensitivity to changes in sets of interrelated parameters and alternative formulations for individual components.

● Maintaining the critical pathway focus in data collection. Models should be an integral part of the decision and allocation process. They provide a framework for identifying needed information, evaluating its contribution to improving our understanding of the system and thereby developing criteria for appropriate allocation of research efforts. Such criteria will differ depending on specific circumstances of a radiological release.

REFERENCES

1. Bates, R. Critical appraisal of current toxicological approaches utilized in context of health risk evaluation. In: Symposium on Health Risk Analysis, Gatlinburg, Tenn., October 27-30, 1980, P. J. Walsh, Ed., Oak Ridge National Laboratory, 1981.
2. U.S. Department of Energy. Environmental Aspects of Commercial Radioactive Waste Management, Department of Energy, Office of Nuclear Waste Management, Washington, D.C. 20545, (Vol. 1, page 3.8.1).
3. Hoffman, F. O., Ed. Proc. Workshop on the Evaluation of Models Used for the Environmental Assessment of Radionuclide Releases. TIC, Oak Ridge, Tenn., CONF-770901, 1978, pp. 73-84, 77.
4. Fletcher, J. F., and Dotson, W. L. HERMES—A Digital Computer Code for Estimating Regional Radiological Effects from the Nuclear Power Industry. USAEC Report HEDL-TME-71-168, NTIS, 1971.
5. Napier, B. A., Kennedy, W. E., and Soldat, J. K. PABLM—A Computer Code for Calculating Accumulated Radiation Dose to Man from Radionuclides in the Environment. PNL-3209, NTIS, 1980.
6. Soldat, J. K., Robinson, N. M., and Baker, D. A. Models and Computer Codes for Evaluating Environmental Radiation Doses. BNWL-1754, NTIS, 1974.
7. Brenchley, D. L., Soldat, J. K., McNeese, J. A., and Watson, E. C. Environmental Assessment Methodology for the Nuclear Fuel Cycle, BNWL-2219, NTIS, 1977.
8. Garner, R. J. Transfer of Radioactive Materials from the Terrestrial Environment to Animals and Man, CRC Press, Cleveland, Ohio, 1972, pp. 15-20.
9. Russell, R. S. Radioactivity in Human Diets. Pergamon Press, New York, 1966, pp. 194, 195, 302.
10. Rogers, L. E., and Rickard, W. H., Ed. Ecology of the 200-Area Plateau: A Status Report. PNL-2253, NTIS, 1977.
11. Napier, B. A., Roswell, R. L., Kennedy, W. E., and Strenge, D. L. ARRRG and FOOD—Computer Programs for Calculating Radiation Dose to Man from Radionuclides in the Environment, PNL-3180, NTIS, 1980.
12. Baker, D. A., Hoenes, G. R., and Soldat, J. K. FOOD—an interactive code to calculate internal radiation doses from contaminated food products. pp. 204-208. In: Proceedings, Conference on Environmental Modeling and Simulation, EPA 600/9-76-016. Cincinnati, Ohio, April 20-22, 1976.
13. International Commission on Radiological Protection. Report of Committee II on Permissible Dose for Internal Radiation, ICRP Publication 2, Pergamon Press, New York, 1959.
14. Cadwell, L. L. Schreckhise, R. G., and Fitzner, R. E. Cesium-137 in coots (*Fulica americana*) on Hanford waste ponds: Contribution to population dose and offsite transport estimates. In: Low-Level Radioactive Waste Management: Proc. Health Physics Soc. 12th Midyear Topical Symposium. Williamsburg, Va., 1979.
15. Mo, T., and Lowman, F. G. Laboratory experiments on the transfer of plutonium from marine sediments to seawater and to marine organisms. pp. 86-95. In: Radioecology and Energy Resources, C. E. Cushing, Ed., Proc. Fourth National Symposium on Radioecology, Corvallis, Ore., May 12-14, 1975.
16. Adams, L. W., White, G. C., and Peterle, T. J. Tritium kinetics in a freshwater marsh. In: Radioecology and

Energy Resources, C. E. Cushing (Ed.), Proc. Fourth National Symposium on Radioecology, Corvallis, Ore., May 12-14, 1975, pp. 96-102.

17. Vanderploeg, H. A., Booth, R. S., and Clark, F. H. A specific activity and concentration model applied to cesium -137 movement in a eutrophic lake. pp. 164-177. In: Radioecology and Energy Resources, C. E. Cushing, Ed., Proc. Fourth National Symposium on Radioecology, Corvallis, Ore., May 12-14, 1975.

18. Cummings, S. L., Jenkins, J. H., Fendley, T. T., Bankert, L., Bedrosian, P. H., and Porter, C. R. Cesium-137 in white-tailed deer as related to vegetation and soils of the southeastern United States. In: Radionuclides in Ecosystems, D. J. Nelson (Ed.). Proc. Third National Symposium on Radioecology, Oak Ridge, Tenn., May 10-12, 1971, pp. 123-128.

19. Olson, J. S. Equations for cesium transfer in a Liriodendron forest. Health Phys. 11: 1385-1392 (1965).

20. Eberhardt, L. L., and Hanson, W. C. A simulation model for an Arctic food chain. Health Phys. 17: 793-806 (1969).

21. Cataldo, D. A., Garland, T. R., Wildung, R. E., and Thomas, J. M. Foliar absorption of transuranic elements: influence of physiochemical form and environmental factors. J. Environ. Qual. 9: 364-369 (1981).

22. Vaughan, B. E., Wildung, R. E., and Fuquay, J. J. Transport of airborne effluents to man via the food chain. In: Controlling Airborne Effluents from Fuel Cycle Plants, TIC, Oak Ridge, Tenn., CONF-76-0806 1976.

23. Chatfield, E. J. Some studies of the aerosols produced by the combustion or vaporization of plutonium alkali metal mixtures, II. J. Nucl. Mater. 32: 247-267 (1969).

24. Stather, J. W., Howden, S., and Carter, R. F. Method for investigating the metabolism of the transportable fraction of plutonium aerosols. Phys. Med. Biol. 20: 106-124 (1975).

25. U.S. Environmental Protection Agency. National Primary and Secondary Ambient Air Quality Standards. Title 40 Code of Federal Regulations, Part 50, Appendix B: Reference Method for the Determination of Suspended Particulates in the Atmosphere. U.S. Government Printing Office, Washington, D.C., 1979.

26. Panel on Radioactivity in the Marine Environment of the Committee on Oceanography. National Research Council. Radioactivity in the Marine Environment. National Academy of Sciences, Washington, D.C., 1971, pp. 154-159, 247.

27. Wildung, R. E., Routson, R. C., and Schmidt, R. L. Seasonal Changes in Particle Size Distribution, Composition, and Strontium Exchange Capacity of Particulate Matter Suspended in the Columbia River. BNWL-1638, NTIS, 1974.

28. USAEC. Liquid Metal Fast Breeder Reactor Program (Final Environmental Statement), WASH-1535, Vol. 2, NTIS, Springfield, Va., 1974. (air concens., p. 4.3-6, p. 4.4-41; reprocessing, pp. 4.4-12 to 4.4-34; Pu escape, pp. 4.4-46; atmospheric dispersal/deposition, pp. II. G-13 to II.G-14, p. 6-17; soil profile movement, p. II.G-26, food chain estimations, pp. II.G-29 to II.G-32; inhalation and ingestion, pp. II.G-5 and II.G-8).

29. Haller, W. A., and Perkins, R. W. Organic iodine-131 compounds released from a nuclear fuel chemical processing plant. Health Phys. 13: 733-738 (1967).

30. Wildung, R. E., Garland, T. R., and Cataldo, D. A. Environmental processes leading to the presence of organically bound plutonium in plant tissues consumed by animals. In: Proc. International Symposium on Biological Implications of Radionuclides Released from Nuclear Industries. International Atomic Energy Agency, Vienna, IAEA-SM-237/37, in press.

31. Watters, R. L., Edgington, D. N., Hakonson, T. E.,

32. Hanson, W. C., Smith, M. H., Whicker, F. W., and Wildung, R. E. Synthesis of the research literature. In: Transuranic Elements in the Environment, W. C. Hanson, Ed., TID 22800, NTIS, 1980.

33. Energy Research and Development Agency. Final Environmental Statement, Waste Management Operations, Hanford Reservation, Richland, WA. ERDA 1538, Vol. 1 and 2, NTIS, Springfield, Va., 1975. (Chemical Composition: Vol. 2, p. II.1-F-3; Pu Movement Vol. 2, pp. II.1-H-1 to II.1-H-3; Microbial Inhibition: Vol. 2, p. II.1-H-2).

34. Means, J. L., Crerar, D. A., and Duguid, J. O. Migration of radioactive wastes: radionuclide mobilization by complexing agents. Science 200: 1477-1481 (1978).

35. Garland, T. R., Schreckhise, R. G., Wildung, R. E., Cadwell, L. L., McFadden, K. M., and Cataldo, D. A. Environmental behavior and effects of technetium-99 and iodine-129. PNL-3300, pt. 2, NTIS, 1980, pp. 79-81.

36. Wildung, R. E., Garland, T. R., and Cataldo, D. C. Accumulation of technetium by plants. Health Phys. 32: 314-317 (1977).

37. Landan, E. R., Hart, L. J., and Gast, R. G. Uptake and distribution of technetium-99 in plants. In: Biological Implications of Metals in the Environment, H. Drucker and R. E. Wildung, Ed., CONF-750929, NTIS, 1975.

38. Wildung, R. E., McFadden, K. M., and Garland, T. R. Technetium sources and behavior in the environment. J. Environ. Quality 8: 156-161 (1979).

39. Vaughan, B. E., Abel, K. H., Cataldo, D. A., Hales, J. M., Hane, C. E., Rancitelli, L. A., Routson, R. C., Wildung, R. E., and Wolf, E. G. Review of potential impact on health and environmental quality from metals entering the environment as a result of coal utilization. Report to Battelle Energy Program Directors, Battelle Memorial Institute, Columbus, Ohio, 1975.

40. VanHook, R. I., and Shults, W. D., Eds. Effects of Trace Contaminants from Coal Combustion. ERDA 77-64, NTIS, 1977, pp. 23-39.

41. Romney, E. M., Rhoads, W. A., Wallace, A., and Wood, R. A. Persistence of radionuclides in soil, plants and small mammals in areas contaminated with radioactive fallout. In: Radionuclides in Ecosystems, D. J. Nelson, Ed., CONF-710501-P1. NTIS, 1973, pp. 170-176.

42. Bryan, G. W. The occurrence and seasonal variation of trace metals in the scallops, Pecten maximus (L) and Chlamys opercularis (L). J. Marine Biol. Assoc. U.K. 53: 145-166 (1973).

43. Bryan, G. W., and Hammerstone, L. G. Heavy metals in the burrowing bivalve, Scrobicularia plana, from contaminated and uncontaminated estuaries. J. Marine Biol. Assoc. U.K. 58: 401-419 (1978).

44. Bryan, G. W., and Hammerstone, L. G. Adaptations of the polychaete, Nereis diversicolor, to estuarine sediments containing high concentrations of zinc and cadmium. J. Marine Biol. Assoc. U.K. 53: 839-857 (1973).

45. Vaughan, B. E., and Rickard, W. H. Hanford National Environmental Research Park; A Descriptive Summary. PNL-2299, NTIS, 1977, p. 20.

46. Emery, R. M., Klopfer, D. C., and McShane, M. C. The ecological export of plutonium from a reprocessing waste pond. Health Phys. 34: 255-269 (1978).

47. Cataldo, D. A., and Wildung, R. E. 1978. Soil and plant factors influencing the accumulation of heavy metals by plants. Environ. Health Perspect. 27: 149-159 (1978).

48. Wahlgren, M. A., Alberts, J. J., Orlandini, K. A., and Kucera, E. T. A Comparison of the Concentrations of Fallout-Derived Plutonium in a Series of Freshwater Lakes. Radiological and Environmental Research Division Annual Report, January-December 1977, ANL-77-65, Part

III, NTIS, 1977, pp. 92-94.

48. Vaughan, B. E. Pacific Northwest Laboratory Annual Report for 1978, Ecological Sciences. PNL-2850, Pt. 2, NTIS, 1979, pp. 9.15-9.18.

49. Sunda, W., and Guillard, R. R. L. The relationship between cupric ion activity and the toxicity of copper to phytoplankton. J. Marine Res. 34(4): 511-529 (1976).

50. Sunda, W. G., Engel, D. W., and R. M. Thuotte, Effect of chemical speciation on the toxicity of cadmium to grass shrimp, *Palaemonetes pugio*: Importance of free cadmium ion. Environ. Sci. Technol. 12: 409-413 (1978).

51. Schmidt, R. L. Copper in the marine environment, Part II. In: CRC Crit. Rev. in Environ. Control. CRC Press, Cleveland, Ohio, 1978.

52. Bowen, V. T., Wong, K. M., and Noshkin, V. E. ^{239}Pu in and over the Atlantic Ocean. J. Marine Res. 29(1): 1-10 (1971).

53. Beasley, T. M., and Cross, F. A. A review of biokinetic and biological transport of transuranic elements in the marine environment. In: Transuranic Elements in the Environment, W. C. Hanson, Ed., TID 22800, NTIS, 1980.

54. Hoffman, F O., and Baes, C. F., III. A Statistical Analysis of Selected Parameters for Predicting Food Chain Transport and Internal Dose of Radionuclides. NUREG/C R-1004, NTIS, 1979.

55. Vanderploeg, H. A., Parzyck, D. C., Wilcox, W. H., Kercher, J. R., and Kaye, S. V. Bioaccumulation Factors for Radionuclides in Freshwater Biota. ORNL-5002, NTIS, 1975.

56. Rygg, B. Release of Radioactive Materials from Nuclear Power Plants, Report No. 2, Disperson Mechanisms, Transport Paths and Concentration Factors for Radionuclides in the Cooling Water Recipient. ORNL-tr-4210, NTIS, 1974.

57. Templeton, W. L., and Brown, V. M. The relationship between concentrations of calcium, strontium and strontium-90 in wild brown trout, *Salmo trutta L.*, and the concentrations of the stable elements in some waters of the United Kingdom, and the implications in radiological health studies. Int. J. Air Water Pollut. 8: 49-75 (1964).

58. Preston, A., Jefferies, D. J., and Dutton, J. W. R. The concentration of cesium-137 and strontium-90 in the flesh of brown trout taken from rivers and lakes in the British Isles between 1961-1966, the variables determining the concentrations and their use in radiological assessment. Water Resour. 1: 475-496 (1967).

59. Ng, Y. C., Burton, C. A., Thompson, S. E., Tandy, R., Kretner, H. K., and Pratt, M. Prediction of the Maximum Dosage to Man from the Fallout of Nuclear Devices. IV. Handbook for Estimating the Maximum Internal Dose to Man from Radionuclides Released to the Biosphere. UCRL-50163, pt. IV, NTIS, 1968.

60. Templeton, W. L. Radiological aspects of seabed dumping in the deep oceans. In: Proceedings of Third NEA Seminar on Marine Radioecology, Tokyo, Japan, October 1-5, 1979, Nuclear Energy Agency of the Organization for Economic Cooperation and Development, 1980, pp. 23-34.

61. U.S. Nuclear Regulatory Commission. Regulatory Guide 1.109: Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I, Revision 1 USNRC, Washington, D.C., 1977.

62. Soldat, J. K. Modelling of Environmental Pathways and Radiation Doses from Nuclear Facilities. BNWL-SA-3939, NTIS, 1971.

63. Hoffman, F. O., Miller, C. W., Shaeffer, D. L., and Garten, C. T., Jr. Computer codes for the assessment of radionuclides released to the environment. Nucl. Safety 18: 343-354 (1977).

64. Houston, J. R., Strenge, D. L., and Watson, E. C. DACRIN—A Computer Program for Calculating Oregan Dose from Acute or Chronic Radionuclide Inhalation. BNWL-B-389, NTIS, 1974.

65. International Commission on Radiological Protection. The Metabolism of Compounds of Plutonium and Other Actinides, ICRP Pub. 19. Pergamon Press, Oxford, England, 1972.

66. Soldat, J. K., and Harr, R. D. Radiation dose model. In: HERMES—A Digital Computer Code for Estimating Regional Radiological Effects from the Nuclear Power Industry, J. F. Fletcher and W. L. Dotson, HEDL-TM E-71-168, NTIS, 1971, pp. 81-161.

67. U.S. Atomic Energy Commission. The Potential Radiological Implications of Nuclear Facilities in the Upper Mississippi River Basin in the Year 2000. NTIS, 1973, p. X-26.

68. Soldat, J. K. Radiation doses from iodine-129 in the environment. Health Phys. 30: 61-70 (1976).

69. Book, S. A., Garner, R. J., Soldat, J. K., and Bustad, L. K. Thyroidal burdens of ^{129}I from various dietary sources. Health Phys. 32: 143-148 (1977).

70. Soldat, J. K. The relationship between I^{131} concentrations in various environmental samples. Health Phys. 9: 1167-1171 (1963).

71. Cataldo, D. A., and Vaughan, B. E. Interaction of airborne plutonium with plant foliage. In: Transuranic Elements in the Environment, W. C. Hanson, Ed., TID-22800, NTIS, 1980.

72. McPherson, R. B., and Watson, E. C. Environmental Consequences of Postulated Plutonium Releases for Westinghouse, PFDL, Cheswick, Pennsylvania, as a Result of Severe Natural Phenomena. PNL-2984, NTIS, 1979.

73. Anspaugh, L. R., Shinn, J. H., Phelps, P. L., and Kennedy, N. C. Resuspension and redistribution in soils. Health Phys. 29: 571-582 (1975).